

# TEM Analysis of Interfaces in Diffusion-Bonded Silicon Carbide Ceramics Joined Using Metallic Interlayers

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## 1. Introduction

properties and applications of SiC

## 2. Sample preparations used for diffusion bonding

Substrates : SA-Tyrannohex<sup>TM</sup> (SA-THX)

Interlayers : Ti-Mo foil

## 3. Experimental results

TEM and STEM images of substrates (SA-THX)

TEM and STEM images of diffusion bonded samples

## 4. Discussion about the microstructure of the formed phases by diffusion bonding

the orientation relation between the precipitated TiC and Mo-Ti (SS)

## 5. Summary

# SiC is an attractive material

(high-temperature, extreme environment applications)

1. Excellent mechanical properties
2. Good oxidation resistance
3. High thermal stability

# Developed for wide range uses

(not only as a monolithic material, but also in composites)

1. monolithic materials injector applications
2. composites materials combustion liner, nuclear and fusion reactor, turbine engine applications



**However**, geometrical limitations hinder the wide use of SiC. It is difficult to fabricate large, or complex shaped components by Hot Pressing or CVD.

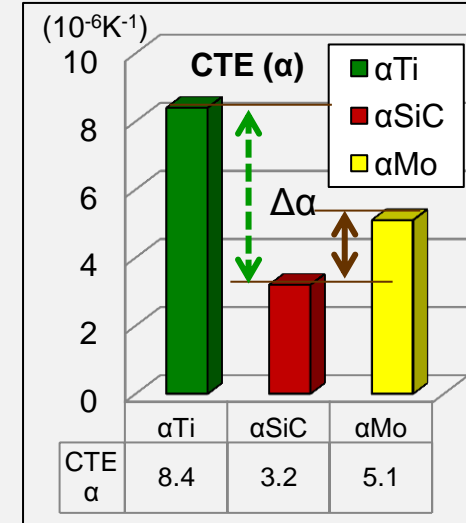
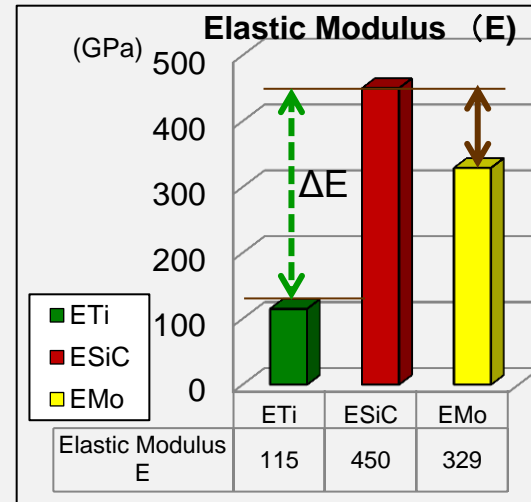
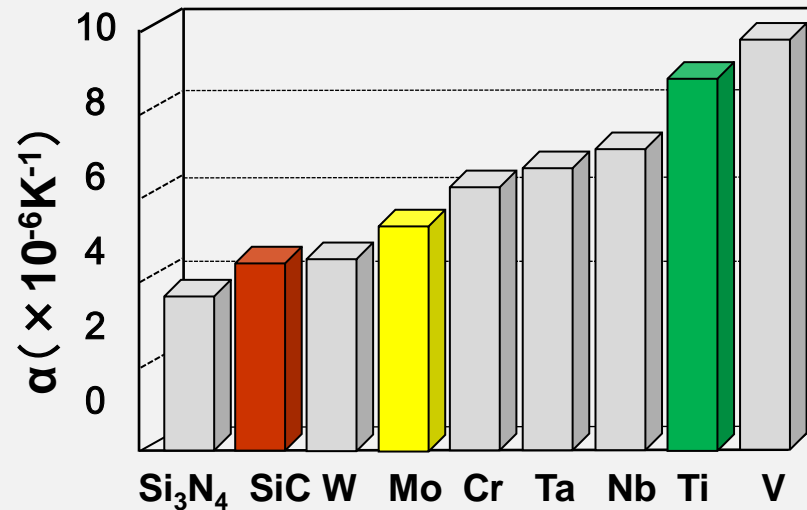
**Therefore, new advanced methods are needed.**

Under those circumstances, one cost-effective solution for fabricating large, complex-shaped components is the joining of simple shaped ceramics.

**In this study, we are going to focus on diffusion bonding.**

# Mismatch of elastic modulus ( $E$ ) and coefficient of thermal expansion (CTE; $\alpha$ ) between substrate and interlayer

We have to pay attention to mismatch of elastic modulus and CTE when we select interlayer material to join SiC.



Both  $E$  and CTE of **Mo** is closer to **SiC** than that of **Ti**.

- Ti and Mo have been used to join  $\alpha$  - SiC.
- Better quality bonds formed with Mo than with Ti.

But,

- Ti can lower the diffusion bonding temperature.

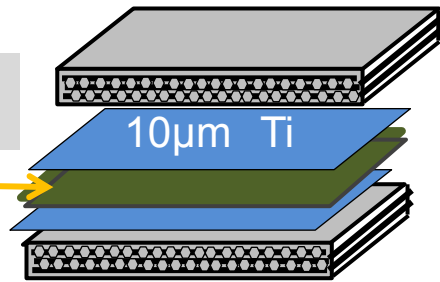
Therefore, **Ti-Mo** bilayer that possesses both advances of Ti and Mo is also very attractive.

Therefore in this work, we utilize Ti-Mo as interlayers.

# Diffusion Bonding of a SA-THX using Ti/Mo metallic Interlayers

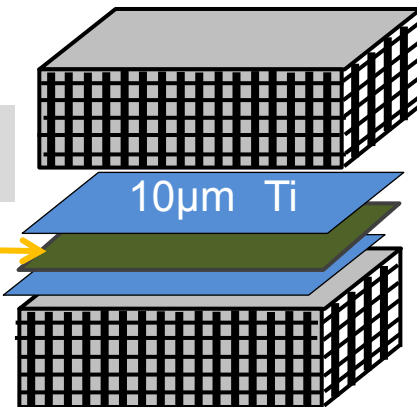
Ti-Mo foil

12.7μm  
Mo



10μm Ti and 12.7 μm Mo  
interlayer  
parallel to SiC fiber

12.7μm  
Mo



10μm Ti and 12.7μm Mo  
interlayer  
Perpendicular to SiC fiber

**Used sample**

@NASA

SA-THX ...SiC fiber-bonded ceramics, UBE Industries

Ti-foil Mo-foil

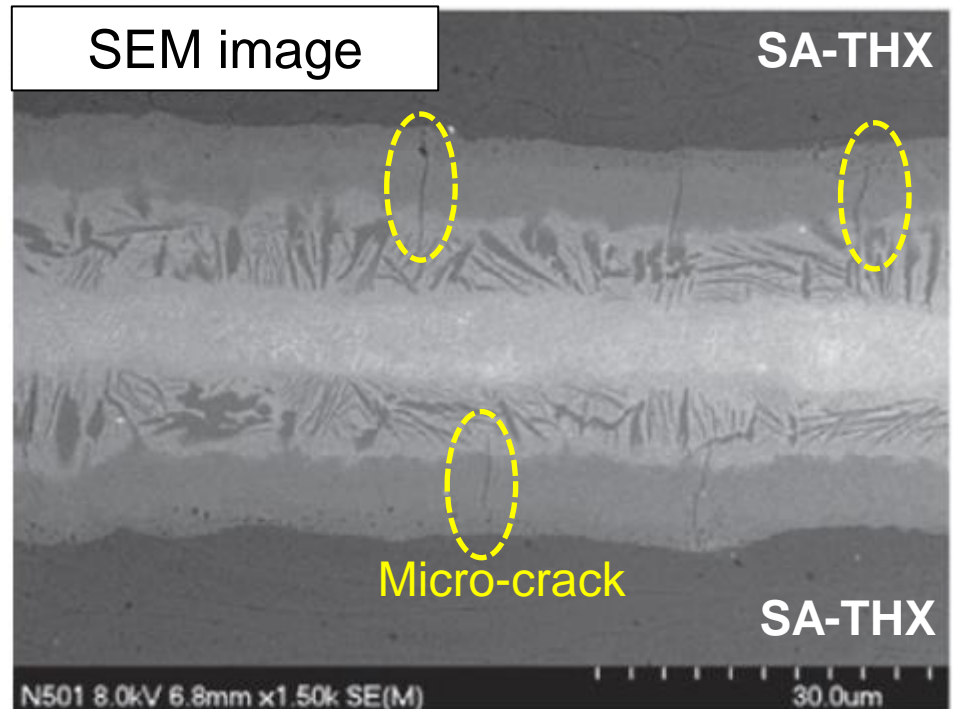
**Bonding structure**

SA-THX // 10μmTi-12.5μmMo-10μmTi // SA-THX

**Bonding process**

Hot-press in 1200°C, 4hour, vacuum 30MPa

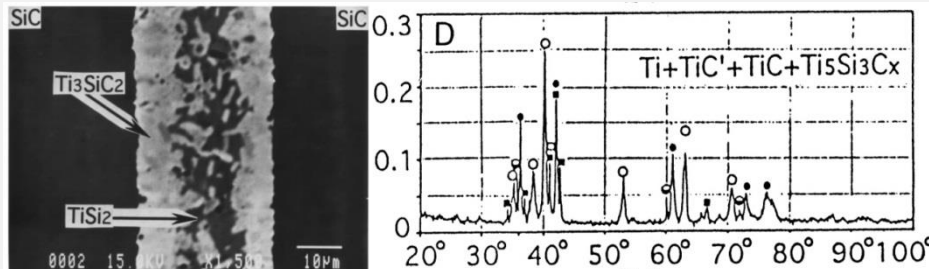
SEM image



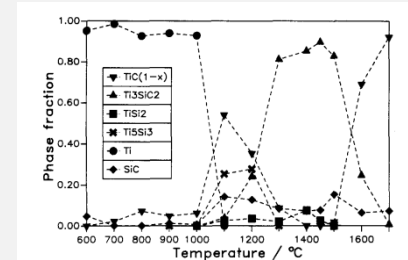
M.C. Halbig, et. al., Ceramics International 41(2015)2140–2149

Until now, the phases formed during diffusion bonding have been studied.....  
(to join SiC-SiC using Ti interlayer)

M. Naka et al, Metallurgical and Materials Transactions, A;  
28A(1997), 1385-1390

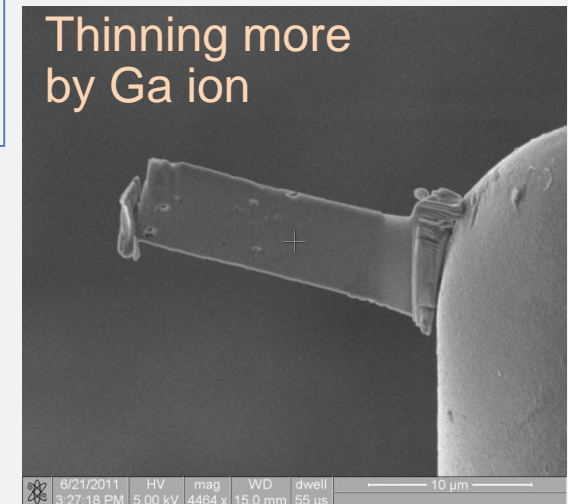
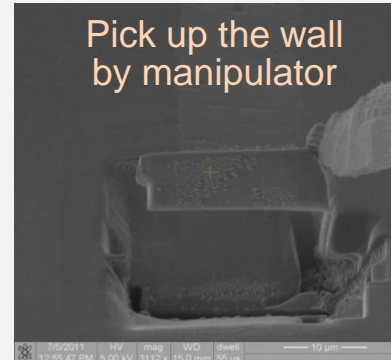
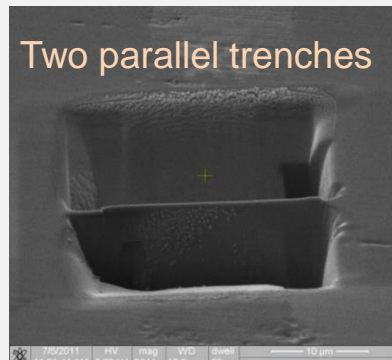
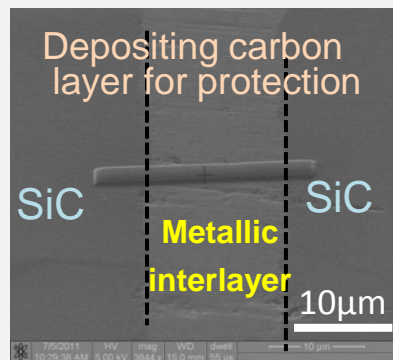


B. Gottselig, et al; J. European  
Ceramic Society, 6(1990), 153-160



Unfortunately, there has been little literature on TEM observation  
of the phases formed during diffusion bonding.

Because, it seems very hard to prepare TEM sample  
from the bonded area. **However**, recently we successfully  
obtained a clean, less-damaged, and precisely selected thin  
specimen from diffusion bonds by using an FIB.



# Objectives

We diffusion bonded SiC and SiC (SA-THX and SA-THX) using Ti-Mo foil metallic interlayer.

We carried out TEM and STEM observations with the diffusion bonded sample prepared by FIB technique.

1. Evaluate microstructures of the diffusion bonded SA-THX by TEM and STEM.
2. Characterize the complex microstructure in the diffusion bonded area by TEM observation and SAED analysis.



# FIB and Cs-corrected STEM

**Focused Ion Beam, FIB**  
(Hitachi FB-2200)



**Cs-corrected STEM**  
(Hitachi HD-2700)



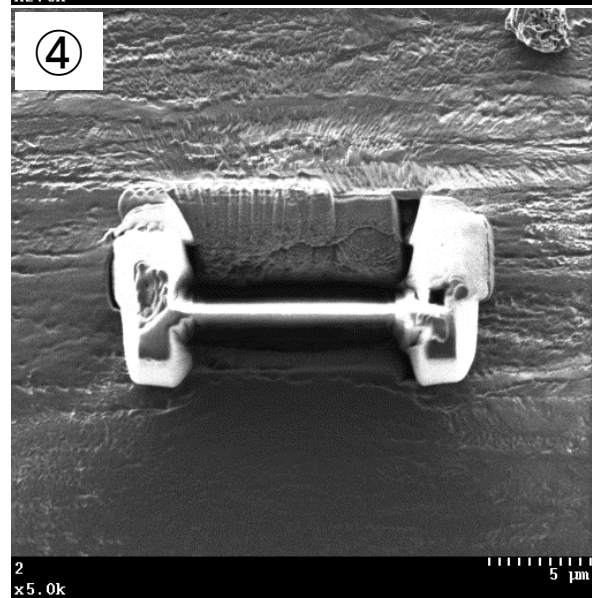
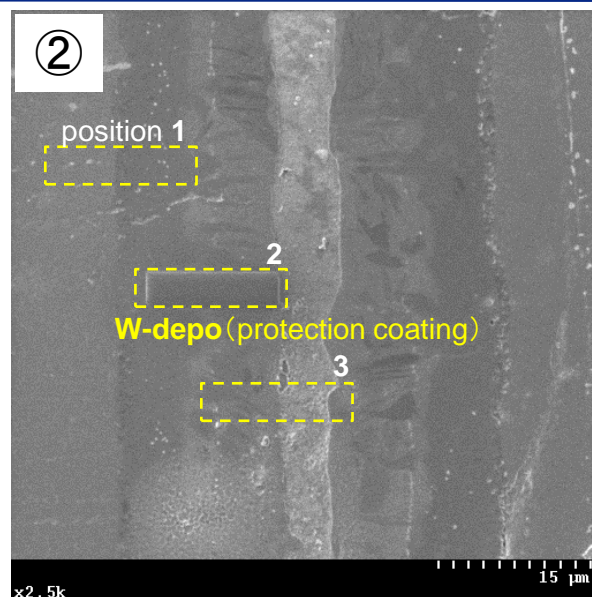
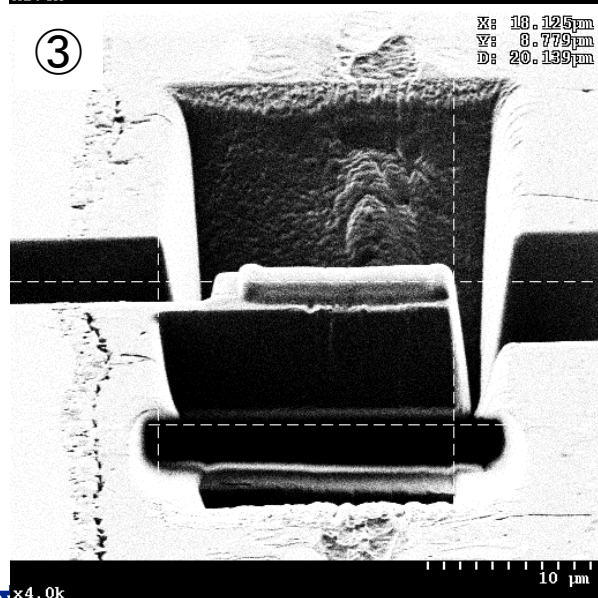
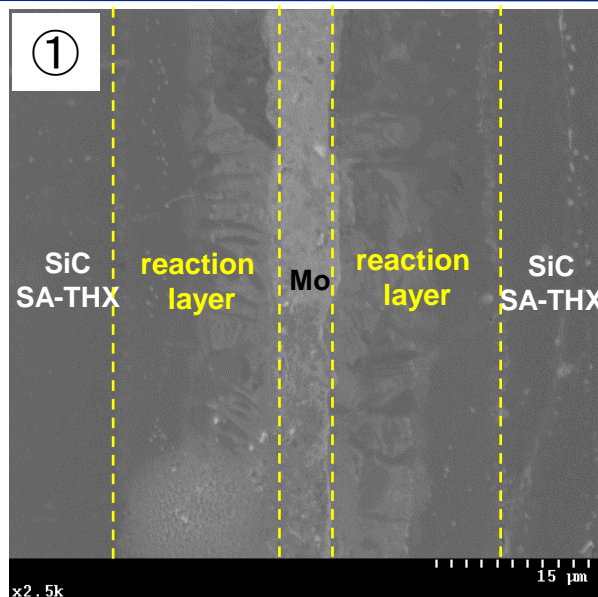
Prepared thin samples for TEM and STEM.

Checked the thin samples prepared by FIB.  
Three-Observation mode:

**SEM, BF-STEM and HAADF**



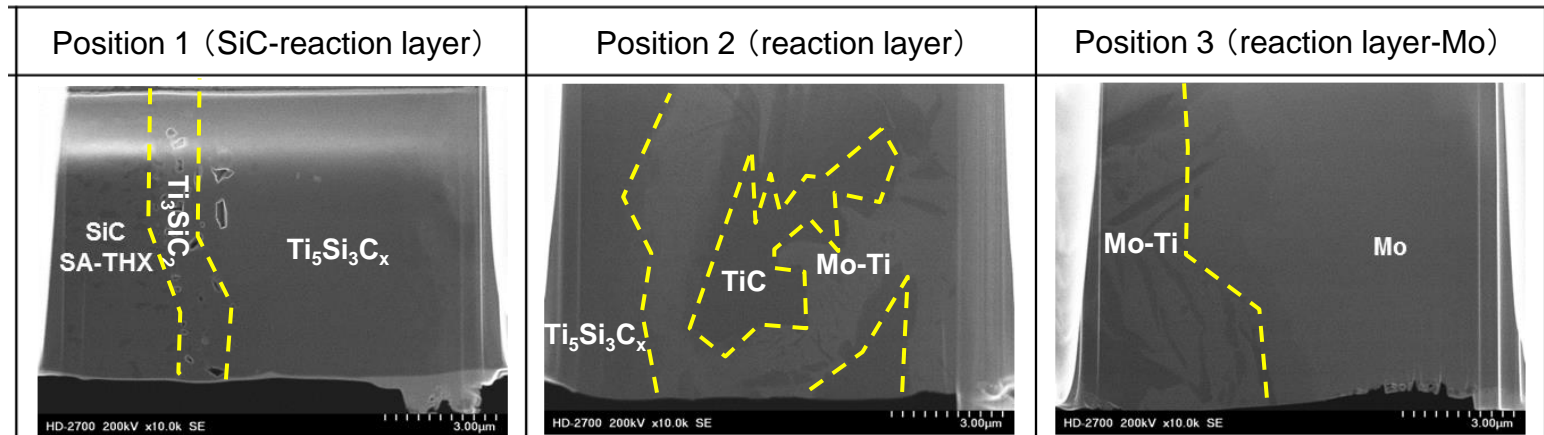
# Fabricating procedure of the thin sample (SIM image obtained by FIB)





# STEM observation of the FIB sample (HD-2700)

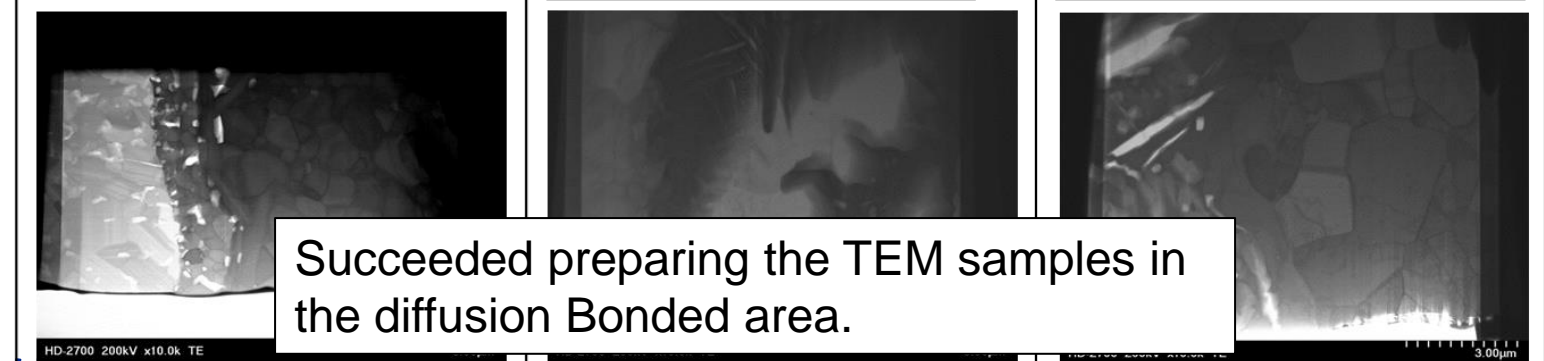
SEM  
image



HAADF  
image



BF-STEM  
image

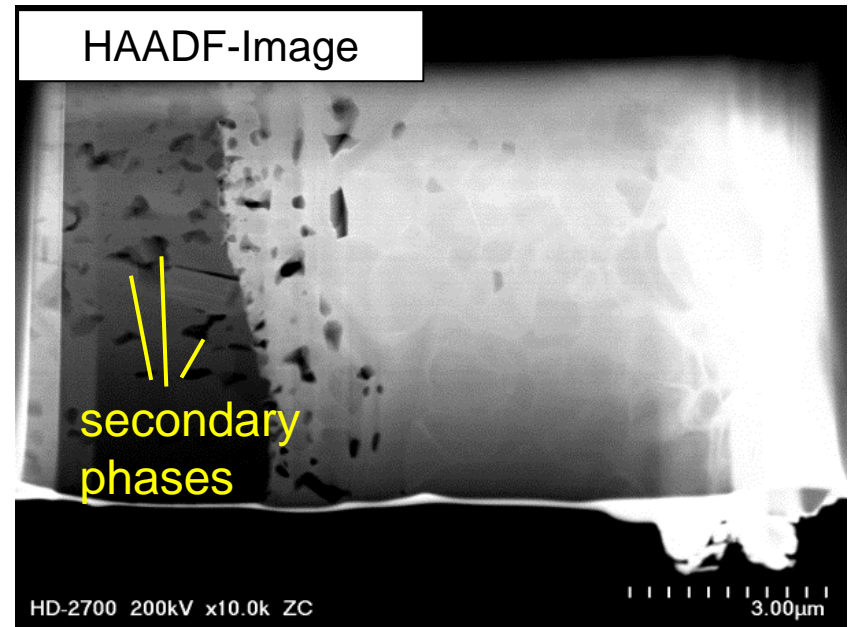
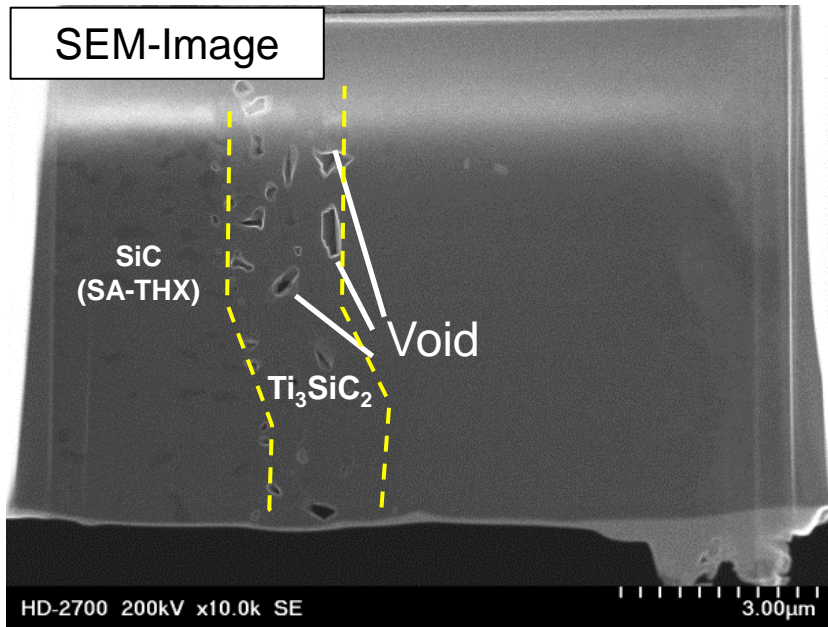


Succeeded preparing the TEM samples in the diffusion Bonded area.



# STEM observation of the FIB sample (HD-2700)

Position 1 (SiC-reaction layer)

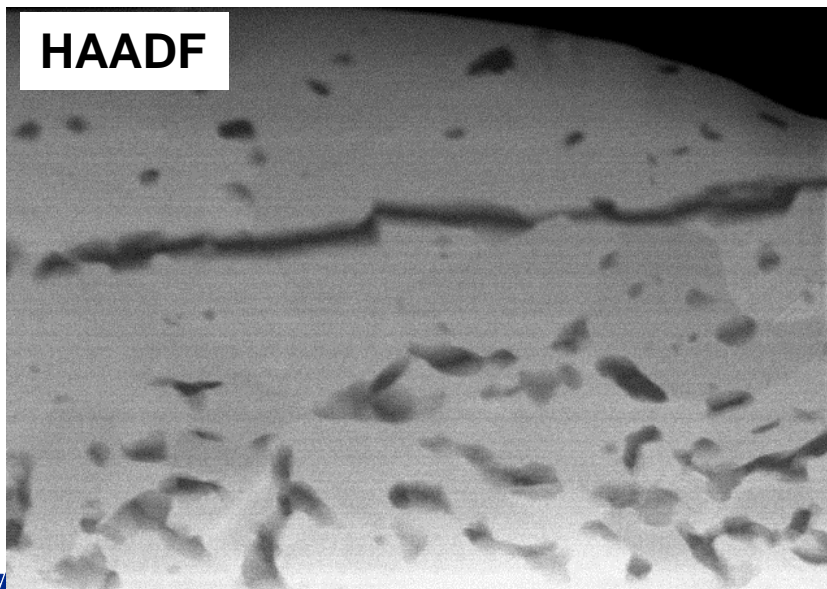
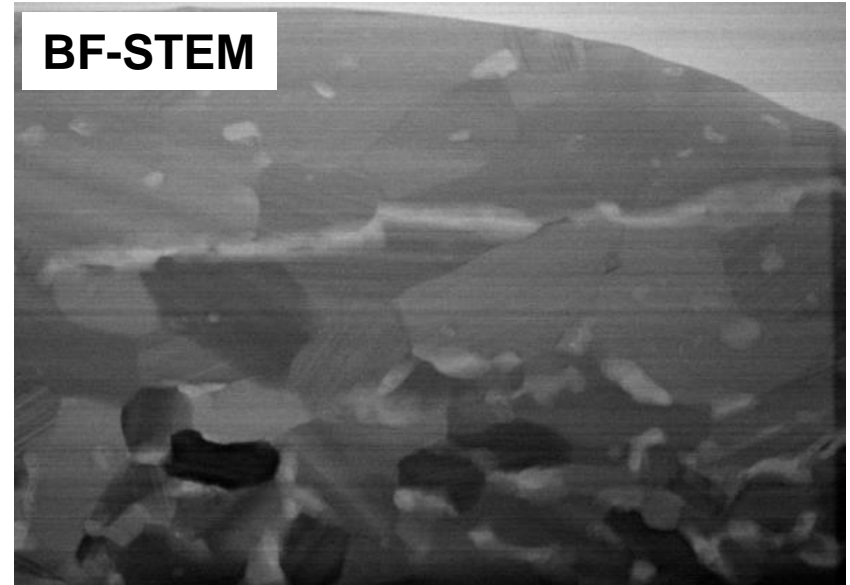
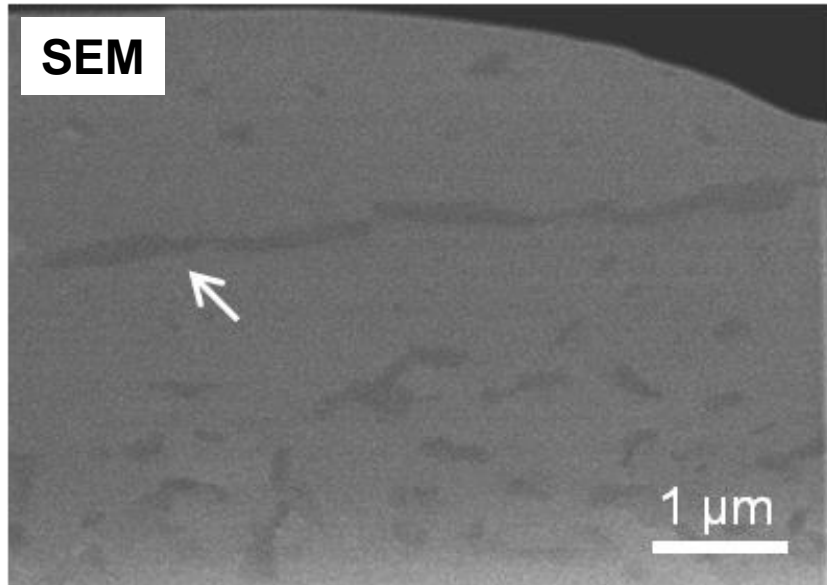


Ti<sub>3</sub>SiC<sub>2</sub> phase ----- Some voids exist.

SA-THX phase ----- Some precipitations (secondary phase) exist.



# STEM images (obtained from SA-THX area.)



Only in HAADF-image,  
the contrast is observed clearly.

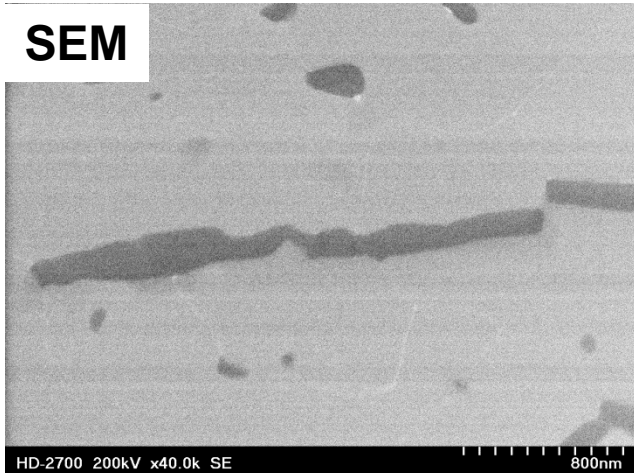
⇒ The precipitations is  
light element.  
**(probably carbon)**



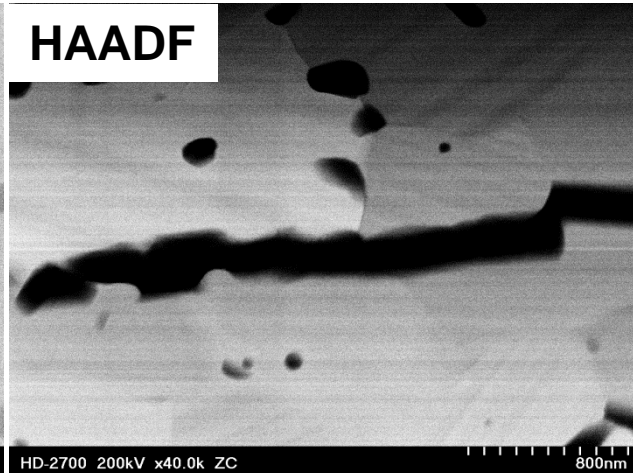
# STEM images (obtained from SA-THX area.)

near the boundary of the SA-THX fiber

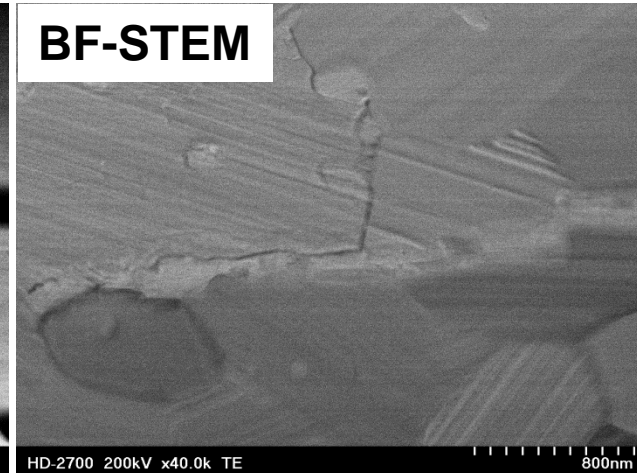
**SEM**



**HAADF**



**BF-STEM**

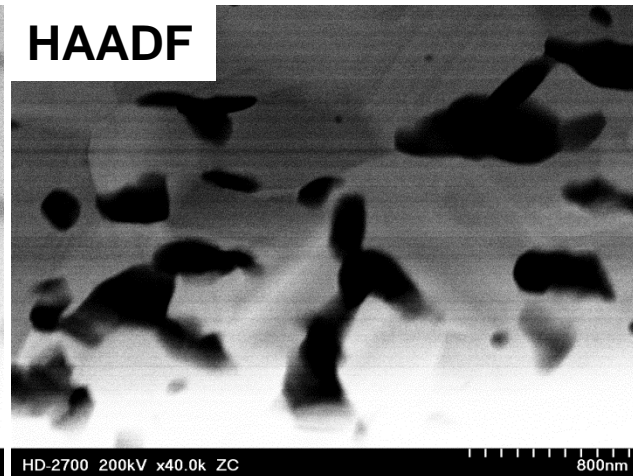


away from the boundary of the SA-THX fiber

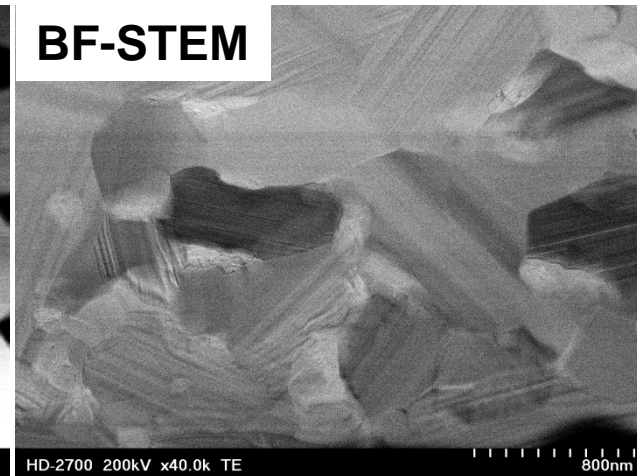
**SEM**



**HAADF**



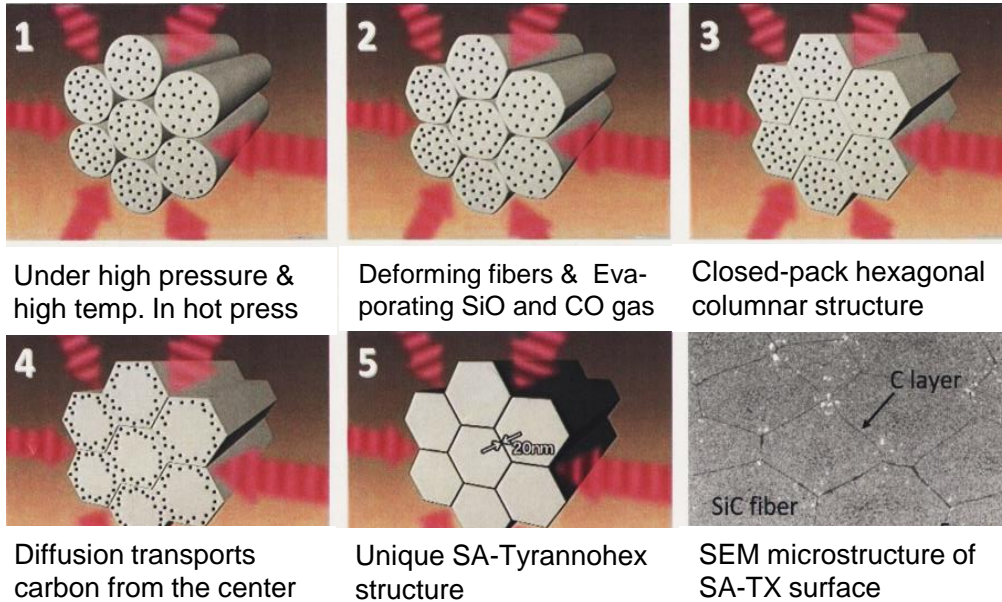
**BF-STEM**





# SA-THX forming process

## SA-THX forming process



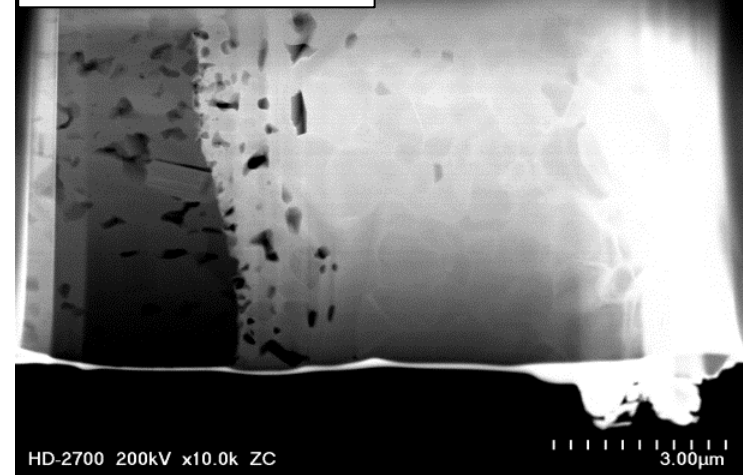
SA-THX has been developed by Dr. T. Ishikawa et al.  
 T. Ishikawa et al, *Science*, 282, 1295-1297 (1998).  
 T. Ishikawa et al, *Nature*, 391, 773-775 (1998).

Also, SA-THX is consisting of a highly ordered, closed-packed structure of very fine hexagonal columnar fibers, with a thin interfacial **carbon layer** between the fibers. The interior of the fiber element was composed of sintered crystalline  $\beta$ -SiC.

⇒ These precipitations stem from residual carbon in SA-THX forming.

## Position 1 (SiC-reaction layer)

### HAADF-Image



The precipitations are not observed in the reaction layer.



The precipitations don't affect diffusion bonding quality a lot?



# Objectives

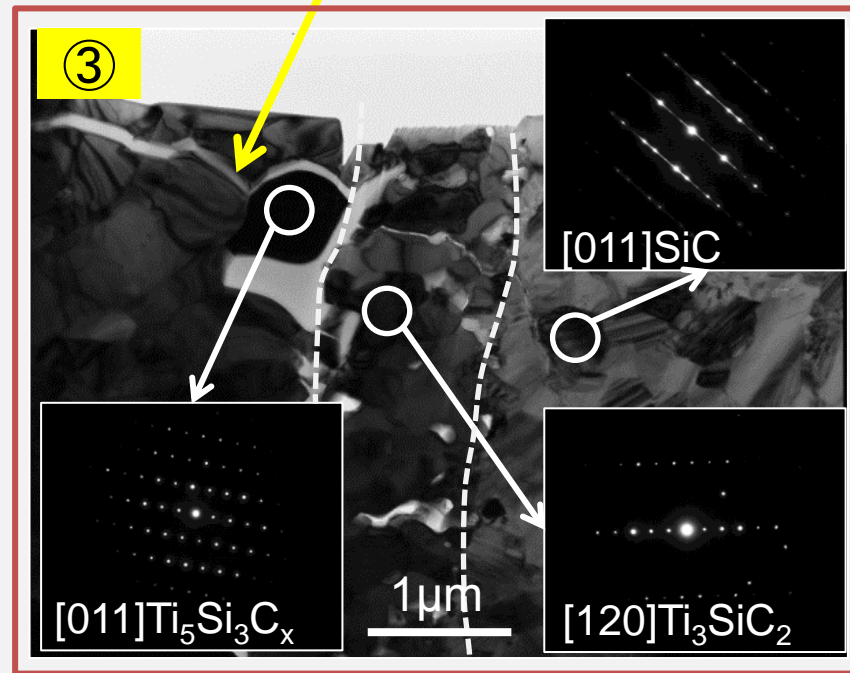
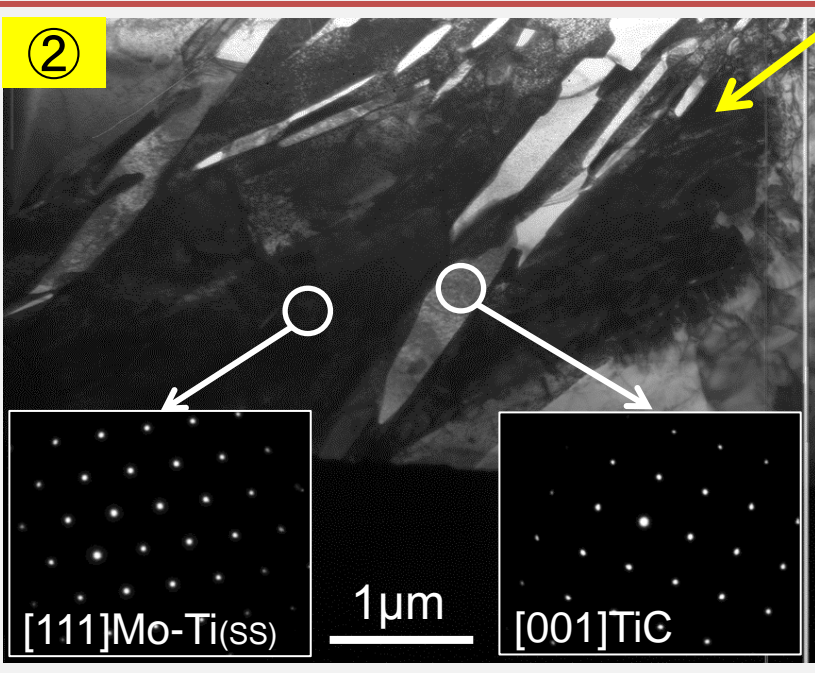
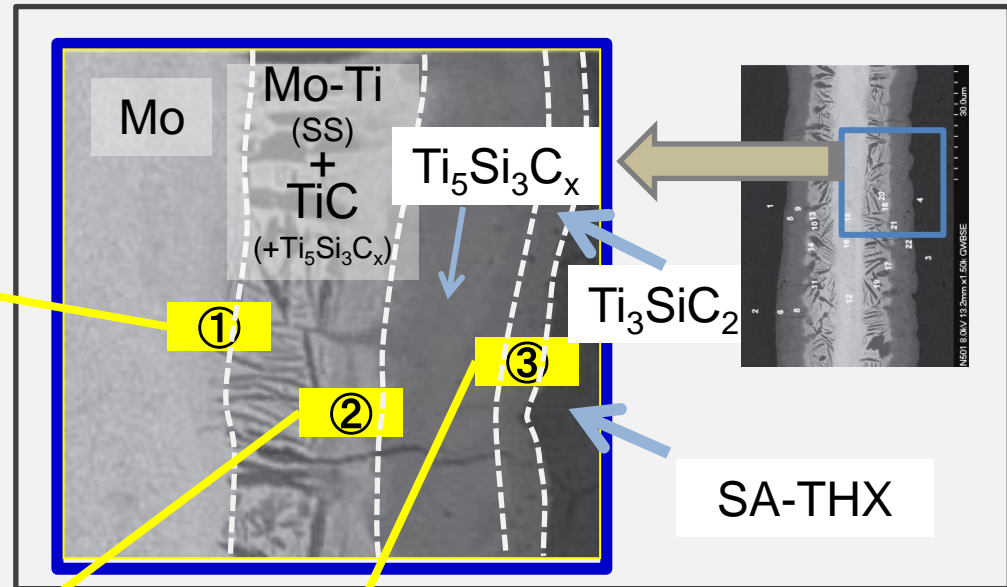
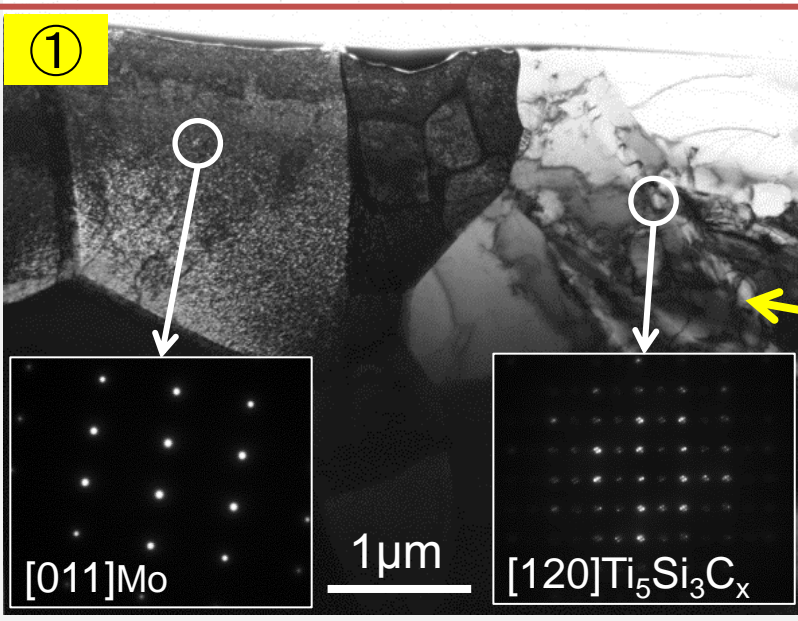
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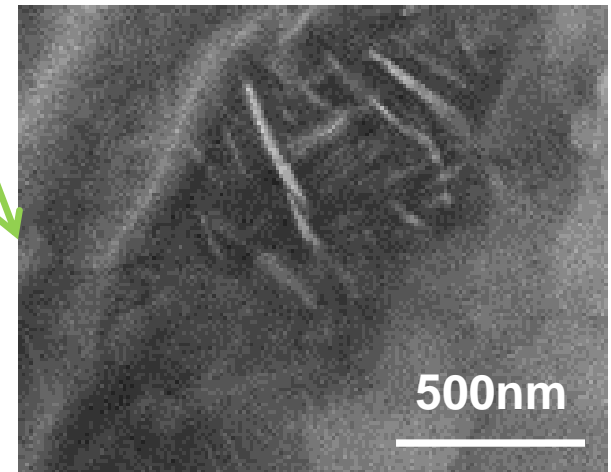
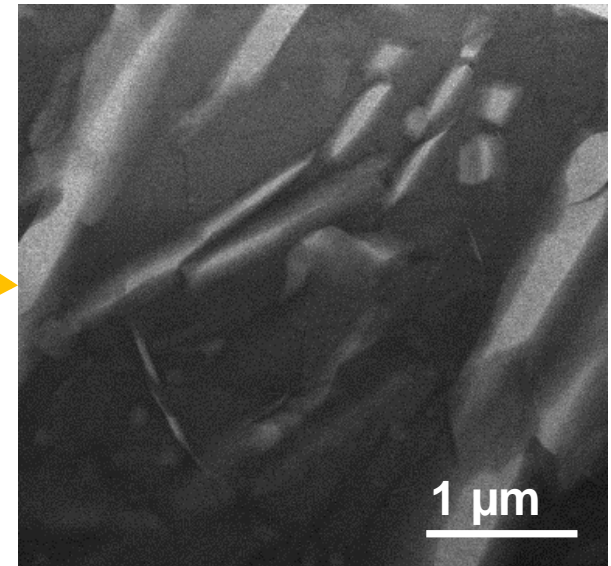
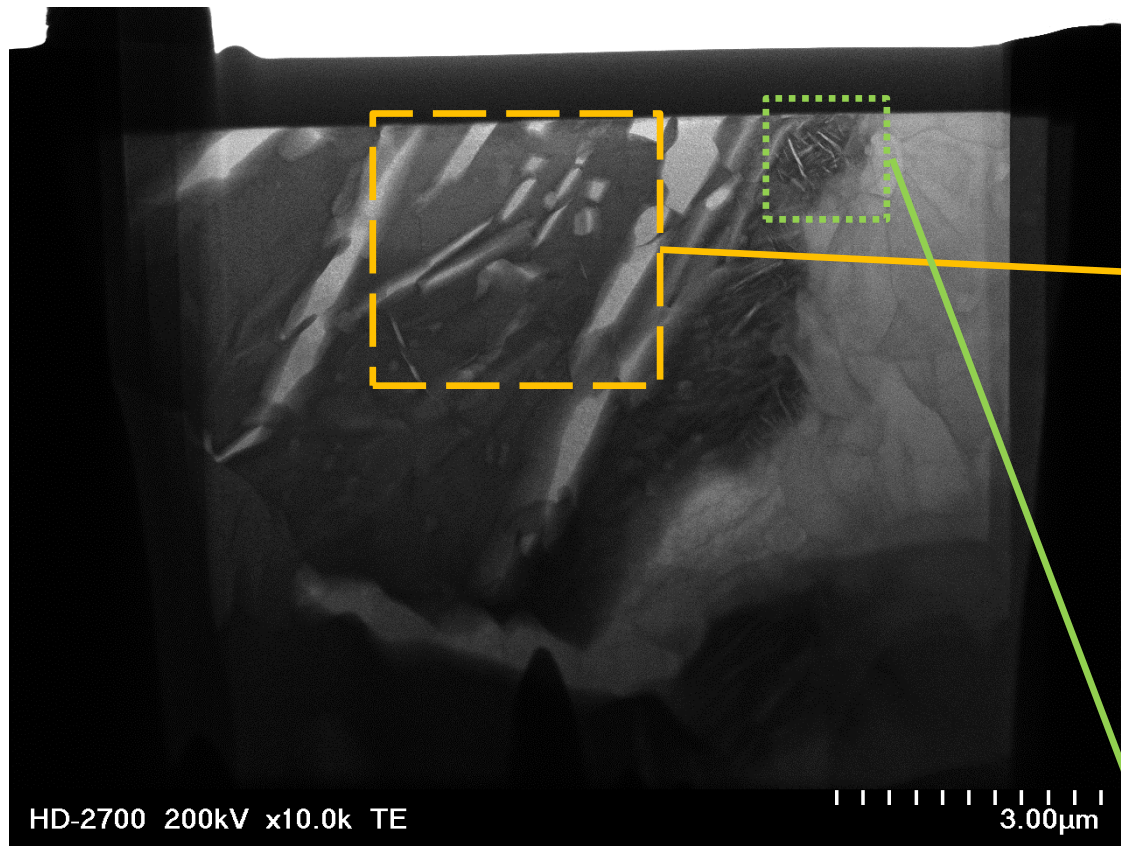
1. Evaluate microstructures of the diffusion bonded SA-THX by TEM and STEM.
2. Characterize the complex microstructure in the diffusion bonded area by TEM observation and SAED analysis.

# TEM image and SAD patterns of diffusion bond (Ti-Mo foil)

@CMCEE11



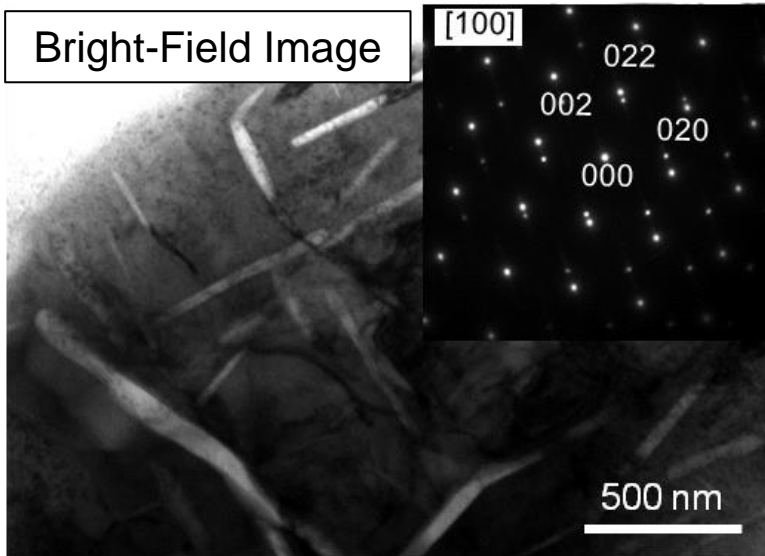
# STEM image of diffusion bond



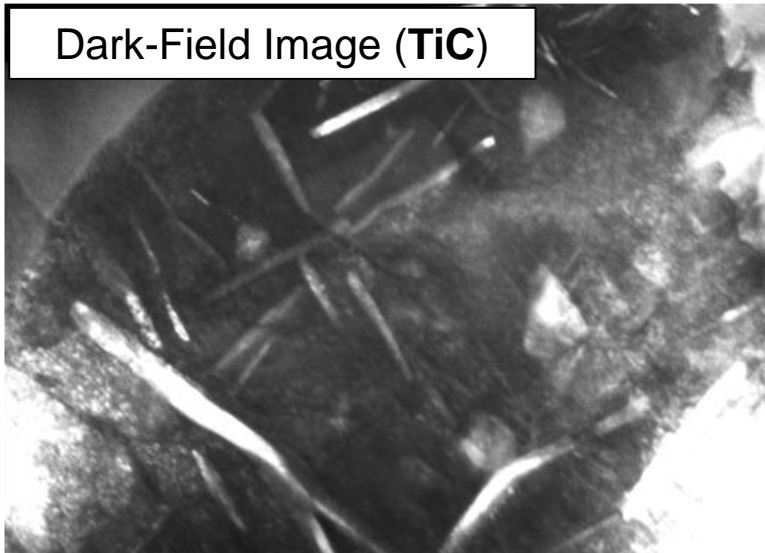
coarse and fine TiC pillars

# TEM image and SAED patterns of TiC pillar

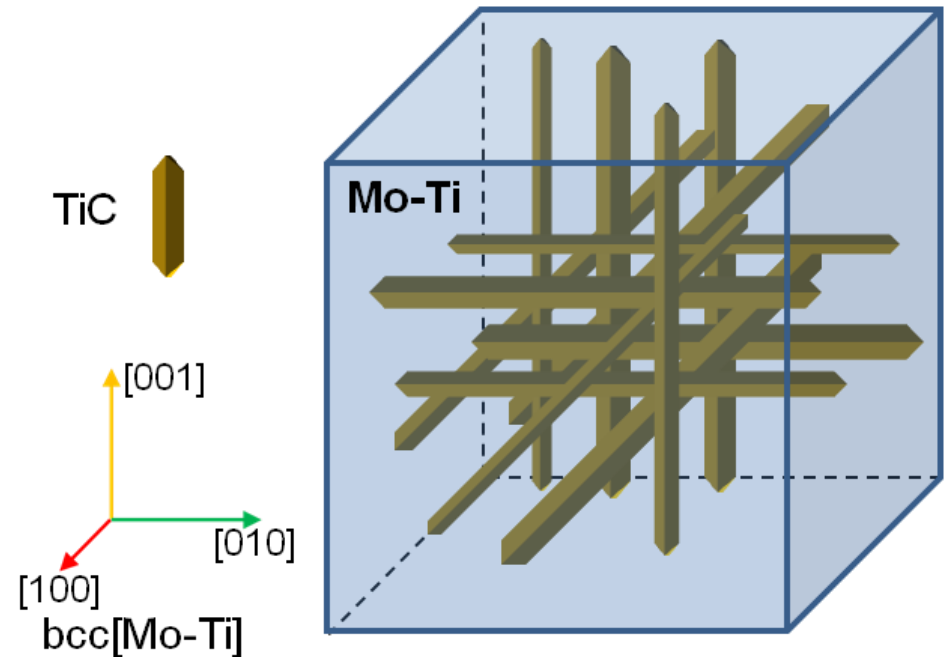
Bright-Field Image



Dark-Field Image (TiC)



Schematic image of the location of TiC in [Mo-Ti]ss matrix.



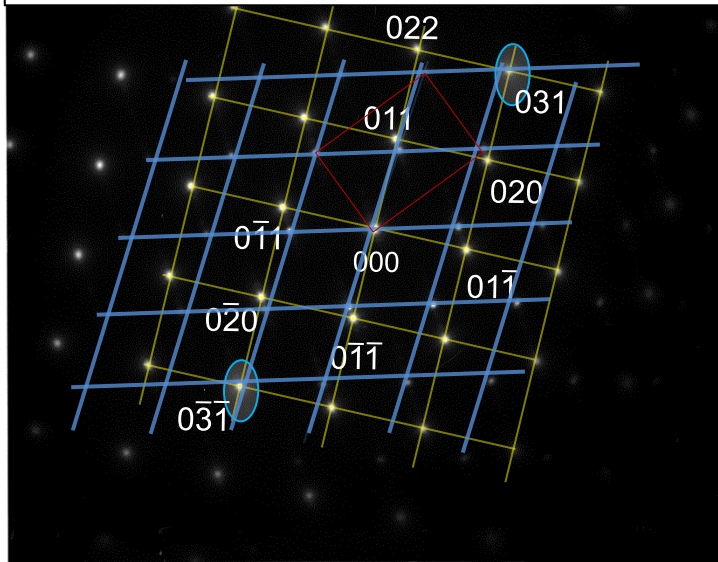
The TiC pillars point to almost  $\langle 100 \rangle$  direction.



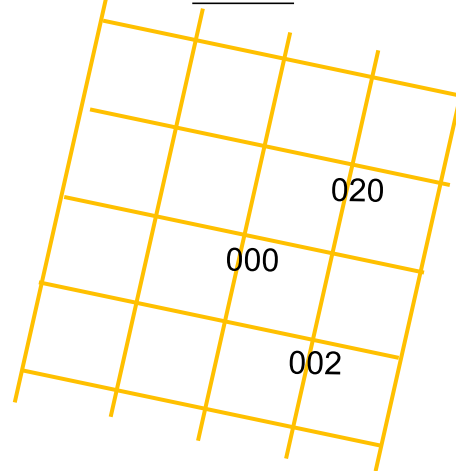


# the relation of the crystallographic orientation between Mo-Ti and TiC

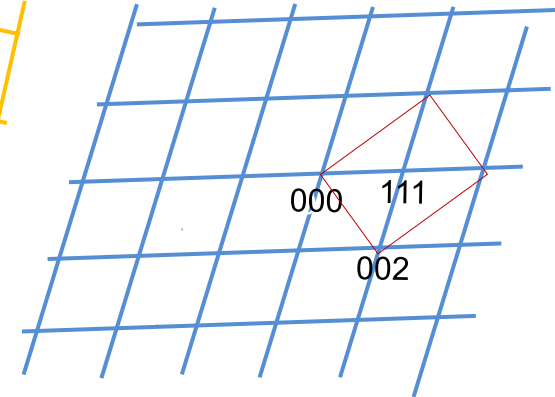
SAED-pattern (Mo-Ti matrix + TiC)



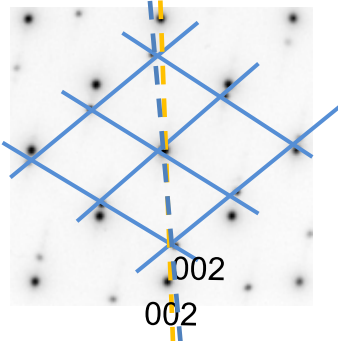
$[100]_{\text{Mo-Ti}}$  bcc-type



$[1-10]_{\text{TiC}}$  NaCl-type

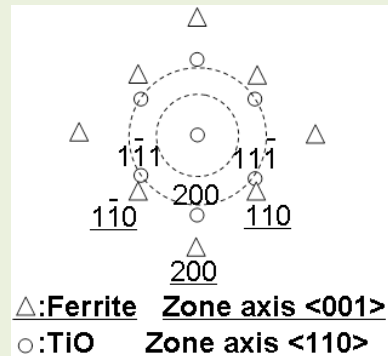


$\langle 001 \rangle$



$[100]_{\text{Mo-Ti}} // [1-10]_{\text{TiC}}$

$(002)_{\text{Mo-Ti}} // (002)_{\text{TiC}}$



**Baker-Nutting's relation**

$[100]_{\text{bcc}} // [011]_{\text{NaCl}}$

$\Rightarrow (002)_{\text{bcc}} // (002)_{\text{NaCl}}$

R.G. Baker and J. Nutting, Precipitation Process in Steels, I.S.I. Special report, No. 64 (1959).

# Summary

- 1. We picked up thin samples from the bonded area of diffusion bonded SA-THX by a FIB micro-sampling technique. The prepared thin samples were sufficiently thin and less-damaged, and allowed the detailed evaluation by TEM and STEM.
- 2. Submicron-sized carbon precipitations were observed in the SA-THX phase away from the boundary of SA-THX fiber. These precipitations did not exist in the reaction phase. It indicates that these precipitations will not affect the diffusion bonding quality a lot.
- 3. TiC pillars were observed around the reaction layer which has a complicated microstructure. The TiC had an orientation relation with the matrix Mo-Ti(SS). In observing from  $[100]_{\text{Mo-Ti}} // [011]_{\text{TiC}}$  incidence, TiC and Mo-Ti were located in almost  $(002)_{\text{Mo-Ti}} // (002)_{\text{TiC}}$  relation. It should be considered that precipitated TiC and matrix Mo-Ti has Baker-Nutting's relation that is often seen when NaCl-type material precipitates in a matrix of bcc-type materials.